Relaxation dynamics of the Holstein polaron

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Keeping the full quantum nature of the problem we compute the relaxation time of the Holstein polaron after it was driven far from the equilibrium by a strong oscillatory pulse. Just after the pulse the polaron's kinetic energy increases and subsequently exhibits relaxation type decrease with simultaneous emission of phonons. In the weak coupling regime partial tunneling of the electron from the polaron self-potential is observed. The inverse relaxation time is for small values of electron-phonon coupling λ linear with λ, while it deviates downwards from the linear regime at $\lambda \gtrsim 0.1/\omega_0$. The imaginary part of the equilibrium self energy shows good agreement with the inverse relaxation time obtained from nonequilibrium simulations.

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Photoexcitation is due to recent technological advances in ultrafast spectroscopy becoming one of the main experimental approaches to disentangle different elementary excitations in a real-time domain on a femtosecond scale. This rapidly developing field enables a novel insight into quantum many-body systems, however it also requires the development of new theoretical concepts. In this context, numerical simulations of nonequilibrium quantum mechanical systems may provide a key insight into processes on a femtosecond time scale.

Several pump-probe experiments reported the observation of self-trapping of excitions emerging after the pump pulse [1–3]. These experiments then triggered theoretical studies of different scenarios of the polaron formation, revealing a complex interplay between a single electron and quantum phonons under nonequilibrium conditions [4, 5].

The role of electron-phonon (e-ph) interaction in several different classes of strongly correlated materials is despite intensive research still ambiguous since a subtle interplay between electron-electron and electron-phonon interaction may lead to various unconventional properties. Pump-probe techniques have shown a potential to identify fingerprints of these interactions during the relaxation process [6–9]. One of the main challenges of such experiments represents the choice of underlying theoretical framework to interpret results. Recently, the phenomenological two temperature model developed long time ago [10, 11], was extended to more involved approaches like the three-temperature model [12] and Boltzmann equation approaches [13–15]. Moreover, the assumption of fast relaxation within the electron subsystem was recently challenged in the case of strong electronic correlations [16]. In contrast to many previous findings it was shown that in the $1D$ strongly correlated system coupled to phonons, the relaxation on short time scales is mostly due to e-ph interaction [17].

Theoretical studies of the polaron motion in strong but constant electric field started with the seminal work by Thornber and Feynman [18]. Later works mostly relied on the rate or Boltzmann equations [19–21]. While quantum coherent effects are absent in the Boltzmann description [22], some recent approaches [23, 24] show that taking fully into account quantum effects is decisive to obtain proper electric field dependence of the drift velocity at large electric fields. Since most past works focused on the influence of a constant electric field on the Holstein polaron, the impact of a short oscillatory pulse on polaron relaxation dynamics remains an open problem despite significant advances in ultrafast spectroscopy [3, 25, 26].

In this Letter we present results of a fully quantum mechanical time evolution of the Holstein model driven far from the equilibrium by a laser pulse. We determine characteristic relaxation times for the system which is initially in equilibrium at zero temperature. The investigated Holstein polaron is subjected to a spatially homogeneous and time-dependent scalar potential that mimics a short laser pulse:

$$\phi(t) = Ae^{-((t-t_c)/t_d)^2}\sin(\omega_p(t-t_c)), \tag{1}$$

which is incorporated into the Hamiltonian via a Peierls substitution in the hopping amplitude:

$$\begin{aligned} H &= -t_0\sum_{l,\sigma}\left[\mathrm{e}^{i\phi(t)}\ c^\dagger_{l,\sigma}c_{l+1,\sigma} + \mathrm{H.c.}\right] + g\sum_j n_j(a^\dagger_j + a_j) \\ &+ \omega_0\sum_j a^\dagger_j a_j, \end{aligned} \tag{2}$$

where $c^\dagger_{l,\sigma}$ and $a^\dagger_i$ represent electron and phonon creation operators at site i, and $n_i = c^\dagger_i c_i$ is the electron density. ω_0 denotes the dispersionless phonon frequency and t_0 is the nearest neighbor hopping amplitude.

The system is described by two dimensionless parameters α and λ where $\alpha = \frac{\omega_0}{t_0}$ and $\lambda = g^2/(2\omega_0 t_0)$ that

determine the crossover from adiabatic($\alpha \ll 1$) to nonadiabatic ($\alpha \gg 1$) limit and the weakly dressed electron ($\lambda \ll 1$) to a heavy polaron ($\lambda \gg 1$). We measure the electric field $F = -\partial_t \phi(t)$ in units of $t_0/e_0 a$, where e_0 is the unit charge and a is the lattice distance. The unit of energy is given by hopping amplitude t_0 and the unit of time is $\hbar/t_0$. From here on we set $a = e_0 = \hbar = t_0 = 1$. Unless explicitly stated the phonon frequency is set to $\omega_0 = 1$. The pulse in Eq. (1) is centered at $t_c = 5$, while the width is given by $t_d = 2$.

We solve the time-dependent Schrödinger equation for a single Holstein polaron on an infinite one-dimensional chain. We use the numerical method based on the exact diagonalization of the variational Hilbert space that led to numerically exact solutions of the polaron ground state [27] and low-lying excited-state properties [28, 29], as well as for description of the time-dependent case [24]. The total energy gain from the external pulse is given by

$$\Delta\langle H(t)\rangle = \int \langle j(t)\rangle F(t) dt, \tag{3}$$

where $j(t) = -\partial_\phi H$ is the current operator. Equation (3) is as well used as an additional time propagation accuracy check, see also Ref.[30].

Before examining the more physically relevant case we analyze the action of a pulse in a form of a delta function $F(t) = -\Phi_0 \delta(t)$ that leads to a simple form of the scalar potential $\phi(t) = \Phi_0 \theta(t)$ where $\theta(t)$ is the Heaviside function. Action with $\phi(t)$ on a free electron state ($\lambda = 0$) at $k = 0$ shifts its kinetic energy from $E_{\rm kin} = -2$ at $t < 0$ to $E_{\rm kin} = -2\cos(\Phi_0)$ when $t > 0$, leaving the electron in an excited, but an eigenstate at $t > 0$. In Fig. 1(a) we show results for $\lambda = 0.1$ and $\Phi_0 = \pi$. Just after the pulse, the increase of the total $\Delta E(t) = \langle H(t)\rangle - \langle H(t = 0)\rangle$, as well as the kinetic energy $\Delta E_{\rm kin}(t)$, reach value $\Delta E \sim \Delta E_{\rm kin} \sim -2\cos(\Phi_0) + 2 = 4$ while the change of the phonon energy $\Delta E_{\rm ph}$ remains close to its value in the polaron ground state, *i.e.* $\Delta E_{\rm ph} \sim 0$. After initial time $t \gtrsim t_i \sim 10$, $\Delta E_{\rm kin}(t)$ exhibits a relaxation type exponential decay towards a constant value $\Delta E_{\rm kin}(t \to \infty)$, meanwhile $\Delta E_{\rm ph}(t)$ increases and the electron-phonon interaction term $\Delta E_{\rm e-ph}(t)$ remains nearly a constant. This dynamics is interpreted as a transfer of the excited electron kinetic energy into phonon excitations. In Fig. 1(b) only $\Delta E_{\rm kin}(t)$ is shown for different choices of pulse amplitude Φ_0. In all cases except for $\Phi_0 = \pi/4$, $\Delta E_{\rm kin}(t)$ decrease exponentially with roughly the same relaxation time. In the case of $\Phi_0 = \pi/4$ the increase of $\Delta E_{\rm kin}(t)$ is lower than ω_0 indicating that the energy transfer from the pulse was insufficient to allow electron relaxation via phonon emission, thus no relaxation is observed.

We next consider a more realistic form of a pulse described by the scalar potential of Eq. (1). At small $\lambda = 0.1$ we observe a gain of the total energy $\Delta E(t)$, see Fig. 1(c), signaling that despite rather weak λ, the system has absorbed a substantial amount of energy. Note that after the pulse is switched off for $t \gtrsim 9 = t_{\rm off}$, the total energy remains constant while there is a clear redistribution between expectation values of the parts of the Hamiltonian. Redistribution between the kinetic $\Delta E_{\rm kin}(t)$ and the phonon $\Delta E_{\rm ph}(t)$ part of the total energy clearly indicates a relaxation of the system. After the pulse, $\Delta E_{\rm ph}(t)$ increases with time and $\Delta E_{\rm kin}(t)$ exponentially decreases, while $\Delta E_{\rm e-ph}(t)$ oscillates roughly around zero. This behavior is similar to the case when $\phi(t) = \Phi_0 \theta(t)$, see Fig. 1(a). For $t \gg t_{\rm off}$ most of the gained energy is absorbed by the lattice. We should remark that the relaxed expectation values of the kinetic energy are not the same as before the pulse. As a general rule we find in all other cases $\Delta E_{\rm kin}(t \to \infty) \lesssim \omega_0$, which indicates that a finite value of $\Delta E_{\rm kin}(t \to \infty)$ is a consequence of the gap for optical phonons [22]. Nevertheless, we detect a clear tendency of the exponential decay of the kinetic energy towards only a slightly elevated value $\Delta E_{\rm kin}(t \to \infty)$ in comparison to the initial energy $E_{\rm kin}(t = 0)$. We have also computed the relaxation of the kinetic energy for different amplitudes of the pulse, presented in Fig. 1(d). We found that the decay time is within our numerical accuracy independent of the amplitude of the pulse A as long as A exceeds a threshold value $A \gtrsim 1$, see also the inset of Fig. 1(d), which is in agreement with the Boltzmann theory.

Our results are qualitatively consistent with experiments on quasi-one-dimensional systems [31], where exponential relaxation takes place [1, 2]. Relaxation was also observed in $2D$ systems [26, 32, 33], where it is a consequence of the molecular dipoles rearrangement in the vicinity of the electron, which can be described by formally equivalent electronic polaron model [34].

In the intermediate coupling regime, namely $\lambda = 1.0$, a different response of the system is expected due to a bigger gap between the polaron band and the continuum of excited states. After the pulse almost the entire excess energy is absorbed into the lattice vibrations, *i.e.* $\Delta E(t > t_{\rm off}) \sim \Delta E_{\rm ph}(t > t_{\rm off})$, see Fig. 1(e). In addition to a large increase, the latter displays as well oscillations with the period corresponding roughly to the phonon frequency. In Fig. 1(f) we present results of $\Delta E_{\rm kin}(t)$ for different amplitudes of the pulse. Apart from pronounced oscillations, we again observe approximately exponential decay in the kinetic energy, roughly independent of the strength of A when $A \gtrsim 1$. We also observe distinct long-time limits of kinetic energies, however in all cases $\Delta E_{\rm kin}(t \gg t_{\rm off}) < \omega_0$.

To get further insight into the relaxation dynamics we calculated the average number of phonon quanta located at a given distance x from the electron

$$\gamma(x) = \langle \sum_i n_i a^\dagger_{i+x} a_{i+x} \rangle, \tag{4}$$

satisfying the sum rule $\langle n_{ph}\rangle = \sum_x \gamma(x)$. At time $t =$

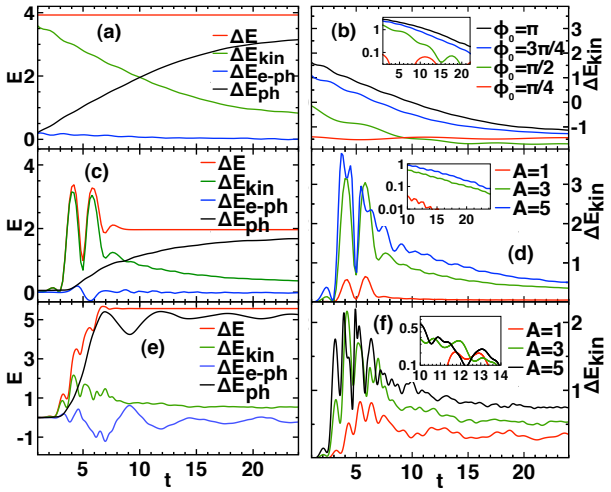

FIG. 1: (a) Expectation values of different parts of the Hamiltonian vs time at $\lambda = 0.1$ and $\omega_0 = 1.0$, after action of $\phi(t) = \Phi_0\theta(t)$, for $\Phi_0 = \pi$. (b) $\Delta E_{\rm kin}(t)$ vs t for different $\Phi_0 = \pi, 3\pi/4, \pi/2, \pi/4$, while other parameters are the same as in (a). (c) Expectation values of different parts of the Hamiltonian vs time at $\lambda = 0.1$ and $\omega_0 = 1$, after action of pulse as given in Eq. (1) with $A = 3$, where $\omega_p = 1.5$ is set close to the maximum of the optical conductivity, while parameters $t_c = 5$ and $t_p = 2$ remain unchanged throughout this work. (d) $\Delta E_{\rm kin}(t)$ vs t for different values of $A = 1, 3, 5$, the rest is the same as in (c). The inset represent the difference between kinetic energy and $E_{kin}(t \to \infty)$ for the same pulse amplitudes as the main figure. (e) Energy of the different parts of the Hamiltonian vs t for $\lambda = 1.0$ and $\omega_p = 2.5$. (f) $\Delta E_{\rm kin}(t)$ vs t for different values of $A = 1, 3, 5$. while the rest is the same as in (e).

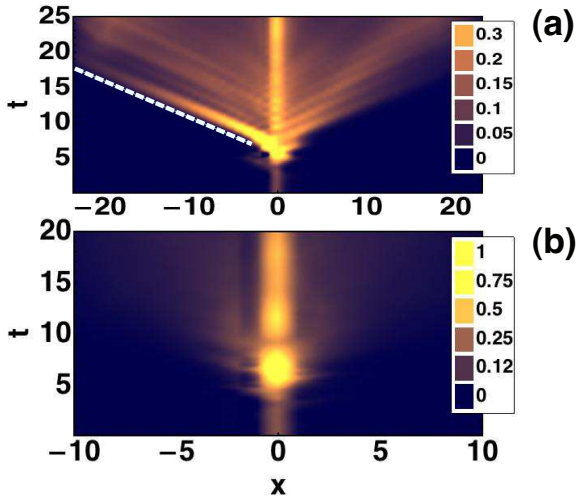

FIG. 2: Electron-phonon correlation function $\gamma(x)$ for pulse amplitude $A = 3$; (a) $\lambda = 0.1$ and the pulse frequency $\omega_p = 1.5$ and (b) $\lambda = 1.0$ and $\omega_p = 2.5$, at different times. Note that there are different scales on both figures.

0, $\gamma(x)$ displays a pronounced peak at the position of the electron ($x = 0$), consistent with the shape of the polaron in equilibrium. In the weak coupling regime, at $\lambda = 0.1$ and $t > t_{\rm off}$, $\gamma(x)$ shows beside original polaron correlation peak also pronounced peaks separating from the central peak in both directions as time increases, see Fig. 2(a). This result is consistent with a hypothesis that a strong pulse splits the polaron into an excited polaron and a nearly free electron. The wavefunction is a superposition of an excited polaron, responsible for the large value of $\gamma(x)$ at $x = 0$ and a nearly free electron, traveling predominantly in the $x > 0$ direction. The more pronounced signal for $\gamma(x < 0)$ can be interpreted as a partial tunneling of electron part of the wavefunction from polaron self-potential that remains located at $x = 0$. The asymmetry on the parity transformation $x \to -x$ is dynamically induced and can be tuned by choosing different shape of the incoming pulse.

The peak at $x < 0$ starts to diminish with time because: *(i)* the escaped nearly free electron is gradually captured by the lattice and *(ii)* the excited electron redistributes its excess energy into constantly spreading area of excited lattice vibrations giving rise to nearly uniform but elevated values of $\gamma(x)$, clearly seen in Fig. 2(a). Here we should stress that redistribution in the correlation function must be due to electron's motion since phonons are dispersionless. This hypothesis is well supported by the estimation of the velocity of the side peaks $v_p = \Delta x/\Delta t \sim 2$ representing the maximal group velocity of the weakly coupled electron [see dashed line in Fig. 2(a)]. Similar partial tunneling was noticed within the adiabatic limit of the driven Su-Schrieffer-Heeger problem [35]. Above considerations give complementary real space interpretation of related experimental results, where excitations are identified in the frequency domain [1, 2]. In the strong coupling regime, namely $\lambda = 1.0$ as presented in Fig. 2(b), the polaron peak at $\gamma(x \sim 0)$ is preserved, but it broadens with time and we observe no peak due to partial electron tunneling.

We computed the relaxation time by fitting the expectation value of the kinetic energy after the pulse with the simple expression $\Delta E_{\rm kin}(t) = \Delta E_{\rm kin}(t \to \infty) + Be^{-t/\tau}$, where $\Delta E_{\rm kin}(t \to \infty)$ is the kinetic energy after relaxation and τ is the relaxation time, see Fig. 3. The inverse relaxation time $1/\tau$ in the extreme weak coupling $\lambda \leq 0.1$ regime shows linear increase with electron-phonon coupling, consistent with $1/\tau \approx 2\omega_0\lambda$. Considering an emission of a phonon by the excited electron with the kinetic energy above one-phonon threshold using Fermi golden rule yields linear dependence on λ, *i.e.* $1/\tau = 2\omega_0\lambda/\sin(k_f)$, where k_f is the final electron's momentum and the inverse relaxation time is determined by the longest decay time, namely at $k_f = \pi/2$. With increasing λ the inverse relaxation time $1/\tau$ first deviates downwards from the linear λ dependence and then saturates as it becomes comparable to the pulse width.

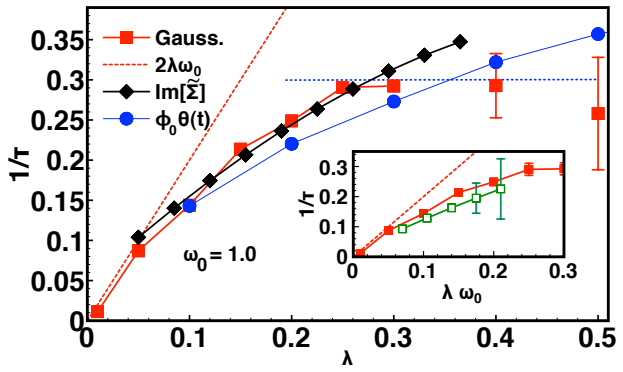

FIG. 3: Inverse relaxation time $1/\tau$ (squares) vs λ for $\omega_0 = 1.0$ using Gaussian pulse, Eq. (1). The dashed line represents $1/\tau = 2\omega_0\lambda$, see discussion in text. Diamonds: the imaginary part of the averaged equilibrium self energy $\mathrm{Im}[\tilde{\Sigma}]$. Circles: $1/\tau$ after the instantaneous pulse $\phi(t) = \Phi_0\theta(t)$ (circles) where pulse strength $\Phi_0 = \pi$ was used. The horizontal dotted line indicates the inverse pulse width. The inset presents $1/\tau$ vs. $\lambda\omega_0$, using Gaussian pulse, Eq. (1), for $\omega_0 = 1$ (full squares) and $\omega_0 = 0.7$ (open squares).

The larger error bars are a consequence of strong oscillations and a smaller spatial extend of the variational phase space in the strong coupling regime. Calculating $1/\tau$ at smaller $\omega_0 = 0.7$ shows that the scaling of $1/\tau \sim f(\lambda\omega_0)$, where $f(x)$ is some unknown function [note that within Fermi golden rule $f(x) = 2x$], persists beyond the linear in λ regime, see the inset of Fig. 3. The saturation of $1/\tau$ is absent in the case when $\phi(t) = \Phi_0\theta(t)$, also presented in Fig. 3. The relaxation time after an instantaneous pulse corresponds to the process averaged over all frequencies and this explains the deviation of the inverse relaxation time from a finite width pulse.

An alternative method for the computation of $1/\tau$ from equilibrium properties is via the imaginary part of the self energy $\mathrm{Im}[\Sigma(\omega)]$ [36] that represents the inverse of the relaxation time of the quasiparticle excitation $1/\tau_\Sigma = \mathrm{Im}[\tilde{\Sigma}(\omega_p)]$. Since $\mathrm{Im}[\Sigma(\omega)]$ depends on the value of the frequency of the pulse ω_p we define $1/\tau_\Sigma$ via the average value $\tilde{\Sigma}(\omega_p) = (1/\sqrt{2\pi\sigma}) \int e^{-(\omega-\omega_p)^2/(2\sigma)^2}\Sigma(\omega)\mathrm{d}\omega$, where $\sigma = 1/t_d$, and integrate over a distribution of frequencies corresponding to the Fourier transform of the pulse. Although there is no a priori reason that this equilibrium quantity represents the inverse relaxation time even after the strong non-equilibrium process the agreement with the actual values of the inverse time is excellent in the weak coupling limit.

To conclude, in this Letter we studied the relaxation dynamics of the Holstein polaron after the strong photoexcitation. In all cases a threshold value of the absorbed energy $\Delta E \sim \omega_0$ exists above which the relaxation dynamics via phonon emission is observed. We computed the relaxation time that is mostly independent of the shape of the pulse. We focused on the values of e-ph coupling λ below the crossover to the strong-coupling regime, *i.e.* $\lambda_c \sim 1$. In this range of λ, relaxation dynamics exhibits two distinct regimes with qualitatively different behavior: the regime of very weak e-ph coupling, $\lambda < 0.1/\omega_0$, and the regime when $\lambda \gtrsim 0.1/\omega_0$.

In the weak-coupling regime $\lambda < 0.1/\omega_0$, $1/\tau$ roughly follows the linear scaling $1/\tau \sim 2\omega_0\lambda$ obtained from the Fermi golden rule. The relaxation process is described by the lowering of the kinetic energy at the expense of the increased phonon energy. A partial tunneling of the electron from the polaron self-potential occurs, resulting in a rapid increase in time of the average distance between the electron and lattice deformation, followed by the subsequent re-trapping process. Moreover, values of τ obtained from real-time calculations can be as well reproduced by calculation of the imaginary part of the averaged electron self-energy.

In the regime of $\lambda \gtrsim 0.1/\omega_0$, $1/\tau$ deviates from the linear in λ dependence well below the crossover to small polaron regime. Most of the energy absorbed by the system is again deposited into phonon excitations, however, in contrast to $\lambda \ll 1$, phonon excitations remain in the close proximity of the polaron. The real-time calculation reveals oscillations in $\Delta E_{\mathrm{kin}}(t)$ and other expectation values, with the period of the phonon frequency, $T \sim 2\pi/\omega_0$. This result is in agreement with a recent study of a half-filled $2D$ Hubbard-Holstein model [37], where oscillations with the period T were observed for $\lambda < \lambda_c$.

In comparison with the experiments our relaxation times are short, which is a consequence of rather high adiabatic coefficient $\alpha = 1$. While in the regime $\lambda \lesssim 0.1/\omega_0$ the effect on τ by lowering α can be obtained from the Fermi golden rule $\tau \propto 1/\lambda\omega_0 = 1/\lambda\alpha t_0$, our numerical results show that even in the regime when $\lambda \gtrsim 0.1/\omega_0$ there exists approximate scaling $\tau \sim 1/f(\lambda\omega_0)$ that can be used to extend our results towards potentially more physically relevant values of α and consequently longer relaxation times τ. Another interesting topic for further research is the effect of the dimensionality, since our simulations agree with the exponential relaxation in quasi $1D$ system, while relaxation is slower in $2D$ experiments.

Let us briefly discuss the possible relevance of our results to correlated electron systems with a finite electron density, such as *e.g.* the Hubbard model. Recent nonequilibrium DMFT study showed that the relaxation time of the pump–excited Hubbard model in the case of large Coulomb repulsion is unexepctedly long [16] due to exponentially slow decay rate of pump-generated doublons, in agreement with experiments on optical lattices [38]. These results open a relevant question about the dominant mechanism of fast relaxation observed in photoexcited strongly correlated materials. Lately, the emission of phonons in the $1D$ Hubbard-Holstein model was indeed shown to be a very efficient relaxation mechanism [17], where a non negligible amount of phonons is already emitted during the application of the pulse. This

observation is in agreement with our results presented in Fig. 1, and suggests that the time-evolution within the full nonequilibrium process [both during and after the pulse] may provide a comprehensive understanding of the photoexcited polaronic systems.

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